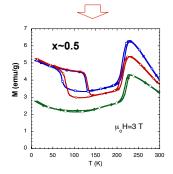
Re-entrant Orbital Order

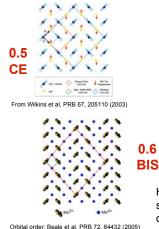
Qing-An Li, H. Claus, K.E. Gray, H. Zheng, S. Klausen, S. Rosenkranz, R. Osborn, D. Mazur and J.F. Mitchell, **Materials Science Division, Argonne National Laboratory**

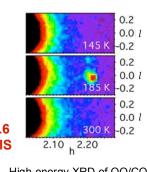
Manganites with x=0.5 (50% hole doping) should exhibit CE-type orbital/charge order (Goodenough). In bilayer manganites, La_{2-2x}Sr_{1+2x}Mn₂O₇, others find re-entrant CE order that is stable at higher temperatures, but not the ground state. Bi-stripe (BIS) orbital/charge order, inferred from the scattering data of others for x=0.6 (60%) is likewise re-entrant.

What have we learned? Magnetization and conductivity data strongly imply that CE orbital/charge order is the ground state, but only in a very narrow range of x, presumably at exactly 0.5.

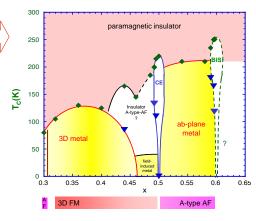
Magnetization identifies x=0.5 crystal with CE order persisting to low temperatures (bottom green data). Conductivity and neutron diffraction data concur.







High-energy XRD of OO/CO superlattice peak, in this case BIS order for x=0.6.



Preliminary phase diagram for bilayer manganites. Compositions near x=0.5 and 0.6 are based on the similarity of ground state properties to near-by x.

Similar trends for x=0.6 suggest the same behavior may exist for the BIS state.

Premise: intense competition with other insulating and metallic phases requires very long-range order and thus nearly stoichiometric x for the orbital/ charge ordered states.

At higher T, CE/BIS order is stable over a wider x-range: consistency with the samples (data) of others.

Open Questions

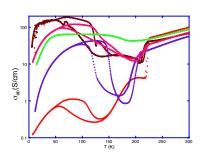
Physical understanding of re-entrance for orbital/charge ordered states?

Are states next to CE order ab-plane metals for x>0.5 and insulators for x<0.5?

What is ground state for precisely x=0.6? What is the magnetic order of the BIS state?

For x~0.46, why are the A-type AF states insulating when theory predicts and data confirm (for x>0.5) that these states are ab-plane metals?

Plans



The variable low-temperature ab-conductivity for crystals made with nominal x=0.5 implies an exquisite sensitivity to exact x.

Establish nature of ground states for x near 0.5 and 0.6.

Multiterminal anisotropic conductivity, magnetization

Tunneling and oxygen k-edge absorption (hybrid states)

X-ray and neutron diffraction

Ideally all on same (small) crystal, checked for homogeneity

Close feedback to crystal growers !!

Address open questions

Explore 0.42<x<0.46

E. Badica, K.E. Gray, J.F. Mitchell and H. Zheng, Phys. Rev. B 70, 174435 (2004)







